ICE NUCLEUS COUNTS AND VARIATIONS AT 3.4 KM. AND NEAR SEA LEVEL IN HAWII

SAUL PRICE

Pacific Supervisory Office, U.S. Weather Bureau, Honolulu, Hawaii

and

JACK C. PALES

Mauna Loa Observatory, U.S. Weather Bureau, Hawaii*

ABSTRACT

Concurrent ice nucleus counts made twice daily at Mauna Loa Observatory (3.4 km.) and near sea level at Hilo, Hawaii, from December 1, 1961, to February 5, 1962, and at intervals of a few minutes during 24 hours on July 10–11, 1963, indicated the wide fluctuations observed elsewhere, with counts varying not only from day to day, but by as much as an order of magnitude within minutes.

Background levels at Mauna Loa Observatory ran well below those at Hilo and averaged under 10 ice crystals in 10 liters at −24°C., values comparable with the lowest reported from other isolated localities. At both sites diurnal variations, evidently reflecting the diurnal mountain circulation and its effects, saw afternoon counts at Mauna Loa Observatory tending to increase with the influx of more turbid air from lower elevations and those at Hilo to decrease from dispersion of the accumulated contaminants by convection and onshore winds.

Although the investigation was not designed to look into fundamental questions, the relatively small distance between the two stations, their large difference in elevation, and the Observatory's isolation for much of the day from the underlying atmosphere permitted inferences concerning such matters as the source and vertical distribution of the ice nuclei and the possibility of extraterrestrial (meteoritic) influences. It would appear that the ice nuclei observed at Hilo did not ordinarily or in major part come from aloft, but rather from the lower atmosphere, and that—in general, and in the presence of the trade inversion—the number of ice nuclei decreased with height in the Hawaiian area.

Major oscillations in the four ice nucleus count-sequences (Mauna Loa Observatory and Hilo, morning and afternoon) appeared to be closely synchronous and to accompany the air mass changes or other synoptic events implied by the deepening and thinning of the moist layer on the Hilo sounding.

1. INTRODUCTION

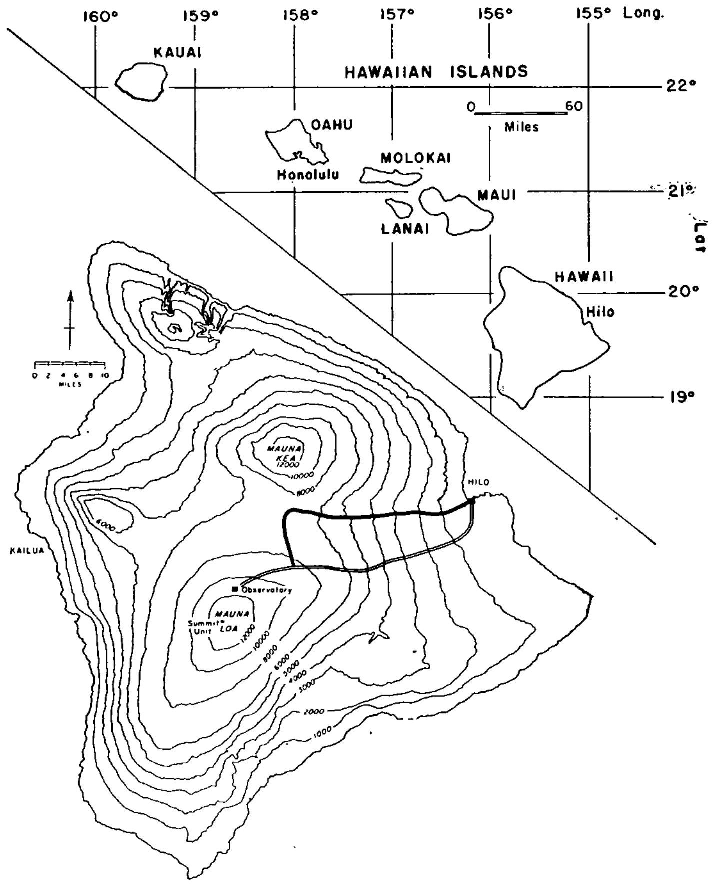
Interest in the sources, nature, and number of the atmospheric ice-forming nuclei stems initially from their possible involvement in precipitation processes of the well-known Findeisen-Bergeron type. Although this interest has prompted much experimental and theoretical work on nucleating mechanisms and on the nucleating efficiencies of various natural and synthetic substances [24], the origin of the naturally occurring ice nuclei and the physical and chemical processes by which they act as nucleating agents remain largely unresolved. However, serial and other observations made in various parts of the world and under diverse meteorological circumstances are helping to define the range and variability of ice nucleus concentrations in nature [5, 19, 21] and some of the local and large-scale atmospheric conditions involved. Recent summaries of the present state of knowledge have been given by Bigg [4] and by Fletcher [16].

The purpose of the present paper is to report on ice nucleus counts made at the Mauna Loa High-Altitude Observatory (MLO) and at Hilo, Hawaii, from December 1, 1961, to February 5, 1962. Although this period is of particular interest to some investigators because it includes a number of the more prominent rainfall singularity dates associated by Bowen [8] with meteor showers, and found also in other geophysical events [3, 11], the authors intend to touch only briefly on this aspect of their data. The importance of these observations is felt to reside chiefly in the sites at which they were obtained: specifically in the inferences made possible by the remoteness of both from sources of industrial pollution and by the relatively small distance and large difference in elevation between them.

2. THE SITES

The Island of Hawaii lies more than 2000 mi. from the nearest continental land mass. It is sparsely inhabited, primarily agricultural (sugar cane and cattle) and with-

*Present address Weather Bureau Research Station, Las Vegas, Nev.



ISLAND OF HAWAII

FIGURE 1.—Map showing Hawaiian Islands and the island of Hawaii with the locations of Hilo and Mauna Loa Observatory.

out significant industry. Air reaching any portion of the island always has a long maritime history, whether it arrives from the sea directly or after some intervening hours over the island. Hilo, on the windward coast ("windward" in Hawaii is always relative to the trades, not to the observed wind) is by far the island's largest community, with a population of 25,000. Ice nucleus counts were obtained at the Hilo airport, within several hundred yards of the sea.

Mauna Loa Observatory, approximately 40 mi. west-southwest of Hilo, and at an elevation of 11,150 ft. (3398 m.), is even more isolated (fig. 1). It is, in addition, frequently insulated during the night and at least part of the day from the lower and more turbid atmosphere by the underlying trade wind inversion, whose height in that area averages some 5000 ft. below that of the station.

These circumstances would appear to make the Observatory especially well suited for baseline determinations of certain of the atmospheric constituents [30, 31], and it is in fact the highest point at which extended serial observations of ice nucleus concentrations have been made.

3. OBSERVATIONS

PROCEDURES

Ice nucleus counts (using the present equipment) have been obtained at intervals at Mauna Loa Observatory since October 1959 [1] and at Hilo since December 1961. The instruments used are identical refrigerated 10-liter expansion chambers of the type described by Warner [35], but modified by the U.S. Weather Bureau (cf. [21]).

In the procedure followed, an air sample is pumped into the chamber and permitted to come into thermal equilibrium with the chamber walls. Rapid decompression then produces a supercooled fog at a temperature predetermined by the initial overpressure. Ice crystals which form in this fog fall into a supercooled sugar solution, where they grow rapidly and may be counted by eye. The chamber is completely purged between successive expansions and counts.

Since the number of nuclei activated is strongly dependent on expansion rate and fog temperature, all counts were made under carefully controlled chamber conditions: expansion ratio, 1.24; wall temperature, -10°C.; fog temperature, -24°C. The latter is well below the threshold temperature of all known, commonly occurring nucleating agents [25, 26] and sufficiently low to activate the smaller and the relatively abundant nuclei of perhaps lower efficiency.

THE DATA

During the period under discussion ice nucleus observations at both sites were made twice daily, at about 08h and 14h (LST). Each observation (frequently referred to hereinafter as a *count*) consisted of the average number of ice crystals counted in five successive purging-expansion cycles.

At Mauna Loa Observatory the long-term background mean thus determined is below 10 nuclei per expansion; i.e., fewer than 1 per liter, a concentration comparable with (and at -24° C. below) the lowest noted over the oceans of the Southern Hemisphere and in other regions remote from human activity [5, 7, 19].

Figure 2 shows the daily morning and afternoon counts at Hilo and MLO from December 1, 1961 to February 5, 1962. The dominant features of the plot—the wide range and variability of the data from day to day and the occurrence of peaks which exceed nearby values by an order of magnitude or more—are those found wherever similar observations have been made [5]. The lesser interdiurnal changes apparently reflect the natural ice nucleus variability of the uncontaminated atmosphere, and have been related to air mass types and air trajectories by some authors, although others have found little connection with meteorological events.

[1] Between December 1957 and October 1959 an earlier model of the same instrument was in use at the Observatory.

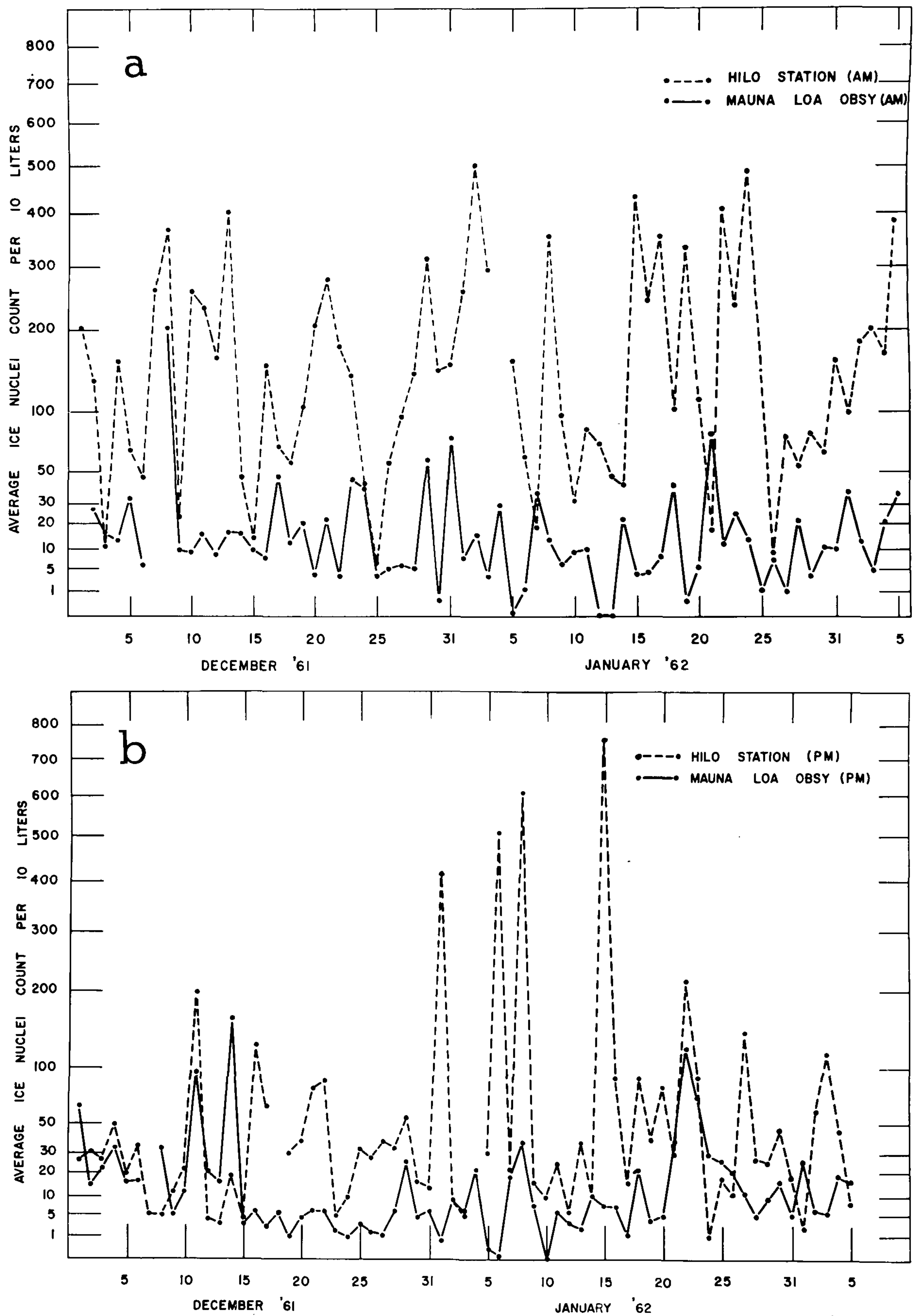


FIGURE 2.—Daily morning (a) and afternoon (b) ice nucleus counts (in 10 liters at −24°C.) at Mauna Loa Observatory and Hilo, December 1, 1961, to February 5, 1962.

TABLE 1.—*Number of ice crystals in 10 liters at −24° C., December 1, 1961, to February 5, 1962**

	Mauna Loa Observatory		Hilo	
	A.m.	P.m.	A.m.	P.m.
Mean	19	18	165	73
Median	11	7	141	28

*Each count was the average of the number of ice crystals counted in five successive determinations (i.e., purging-expansion cycles).

For the peaks there is as yet no systematic explanation other than that of Bowen [8, 10], who attributes them, together with world-wide rainfall singularities, to prior influxes of meteoritic material into the upper atmosphere, an hypothesis still very much in dispute on physical and statistical grounds [12, 13, 16, 22, 33].

It is evident from figure 2 that, on the whole, counts at Hilo are higher and more variable, and peaks more frequent, than at the Observatory. Table 1 confirms that means and medians at Hilo far exceed those at Mauna Loa, the disparity being greater in the morning than in the afternoon.

A more significant dissimilarity between the counts at the two sites and times is strikingly illustrated in the frequency distributions (fig. 3). Thus, at the Observatory most counts were very low, about half having fewer than 10 ice crystals in 10 liters and only 3 more than 100. The Hilo distribution is much flatter, with fewer low values and many more high and very high values than occurred at Mauna Loa.[2]

Further, while at MLO the similarity between morning and afternoon distributions suggests little diurnal variation at that site *during this period* (the subject is discussed at greater length later in this paper), afternoon counts at Hilo range decidedly *below* those made in the morning.

Somewhat obscured by the day-to-day variability, are the extended periods of relatively high or low counts which emerge when the observations are viewed in terms of their respective medians (see fig. 4). Probably the most impressive of these is the 14 successive afternoons of below-median values at MLO during the second half of December—a period with a mean of 3.5, a median of 2.8, and no instances above 6 ice crystals/10 liters. Shorter runs of counts both above and below the median are not uncommon in all four data series, but only the afternoon counts at Mauna Loa, and to a lesser degree the morning counts at Hilo, show a significant interdiurnal persistence (see table 2).

On a number of occasions counts at Hilo were so high on successive mornings, yet so low on the afternoons of the same days, as to give the impression of an "alternation" between a morning regime, characterized by a high

[2] Hilo counts may exceed those at Mauna Loa by even more than these tables and figures indicate. Comparisons made by Kline [18] suggest that the Mauna Loa instrument counts *higher* than does that at Hilo by a factor of about 1.84. On the other hand average air densities at the two sites are in approximately the ratio 2:3. Hence, relative to the Hilo counts, those at MLO are only about two-thirds those given in this paper.

TABLE 2.—*Interdiurnal persistence of direction of departure from the median ice crystal count. On the assumption of a random process, the expectancy for each run length is half the observed frequency of the next shorter run*

		Length of run (days)					
		2	3	4	5	6	7
MLO a.m.	Expected	31.5	15.5	7	3	1	
	Observed	31	14	6	2	0	
MLO p.m.	Expected	32	20	13.5	10	7.5	5.5
	Observed	40	27	20	15	11	9
Hilo a.m.	Expected	31	20	11.5	7	3.5	1.5
	Observed	40	23	14	7	3	0
Hilo p.m.	Expected	30.5	15.5	7.5	3.5	1	
	Observed	31	15	7	2	1	

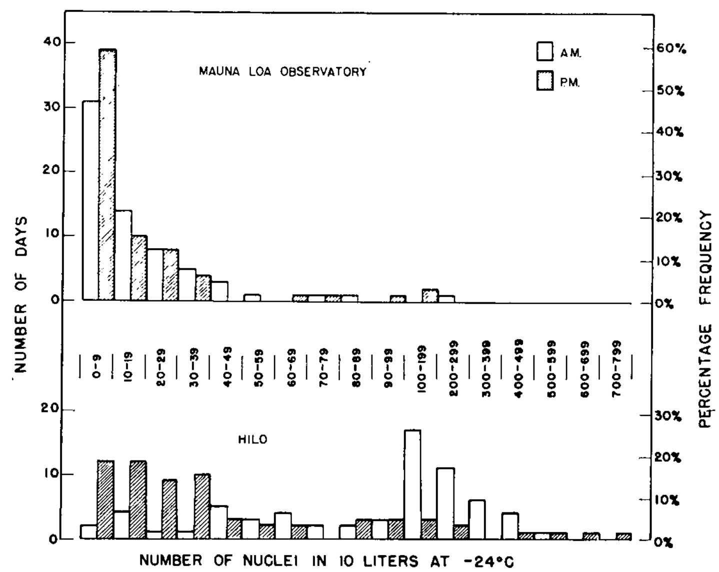


FIGURE 3.—Frequency distribution of morning and afternoon ice nucleus counts at MLO and Hilo (number in 10 liters at −24°C.), showing at Hilo a pronounced diurnal variation and a greater number of higher counts and peaks.

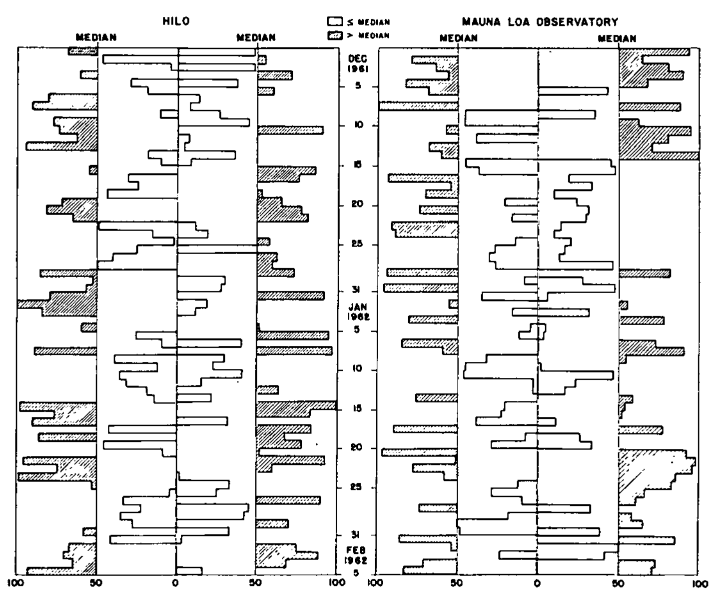


FIGURE 4.—Daily morning and afternoon ice nucleus counts at MLO and Hilo, shown in terms of their respective percentiles. Note in particular the 14 successive afternoons beginning December 14, 1961, during which counts at MLO ran below their median.

ice nucleus content, and a quite different afternoon regime, characterized by a low ice nucleus content. In fact, some of the lowest counts at Hilo occurred on afternoons of days whose morning counts had ranked among the highest: for example, December 7 and 8: a.m., 262 and 368; p.m., 5 and 4 (all counts are ice crystals in 10 liters); December 12 and 13: a.m., 163 and 403; p.m., 4 and 3; January 1 and 2: a.m., 500 and 293; p.m., 9 and 5. Hilo's highest morning count, 488, and the lowest (afternoon) count of the period occurred on the same day (January 24).

4. DISCUSSION AND INTERPRETATION

THE MOUNTAIN CIRCULATION

A number of the important features of the ice nucleus measurements just described can be understood in terms of the meteorological circumstances under which they were obtained. Perhaps the central consideration is that both MLO and Hilo ordinarily lie within an atmospheric envelope generated by the thermal and dynamic influences of Mauna Loa itself on the ambient free air,[3] and having motions and properties often substantially at variance with those of the surrounding atmosphere. Much though a detailed picture of this surface layer and its behavior might aid in the interpretation of the ice nucleus counts made within it, suitable data are too sparse for more than a schematic outline to be drawn.

Observations of temperature, humidity, and wind made at MLO and sketchily elsewhere on Mauna Loa reveal a local circulation strongly diurnal in the classical mode, although somewhat complicated by the trade inversion, which separates the lower cool and moist marine stratum from the dry and warmer air above, and whose height averages only half that of the mountain itself.

[3] 125 mi.^{2} of Mauna Loa's dark lava surface lie above 10,000 ft.

During the night, air radiatively cooled on its upper slopes descends Mauna Loa. It seems likely that adiabatic heating and the intervening inversion bar this current from the lower mountain, and that the downslope flow observed there (e.g., at Hilo) arises independently. These descending currents, which manifest themselves at the Observatory as southerly winds (note the topographic gradients in fig. 1) and possess the low humidity of the neighboring free air, persist until shortly after sunrise, when they are disrupted by local convection and then, within an hour or two, supplanted by the more turbulent daytime wind whose prevailing direction—northerly—is approximately 180° from the nocturnal flow. Although this northerly daytime wind is commonly referred to as "upslope", air reaching the Observatory during the forenoon appears from its low humidity to have originated well above the inversion, and may thus represent chiefly inflow, rather than ascent. Later in the day, however, the dew point at MLO frequently increases, at times quite abruptly, to values which the Hilo soundings identify as characteristic of marine air from below the trade inversion. A typical hygrothermograph is shown in figure 5.

It is not clear whether this moist influx implies primarily an upslope seepage from below an inversion breached by the mountain itself, or that the inversion as a whole has bowed upward sufficiently to bring MLO into the sub-inversion layer; but it appears on some days (from the leveling-off in dew point at MLO) that air reaching the Observatory throughout the afternoon may be drawn largely from immediately below the inversion, rather than from progressively lower depths.

In late afternoon the daytime winds begin to fail, and sunset is followed within about an hour by a resumption of the downslope flow. The morning and evening transi-

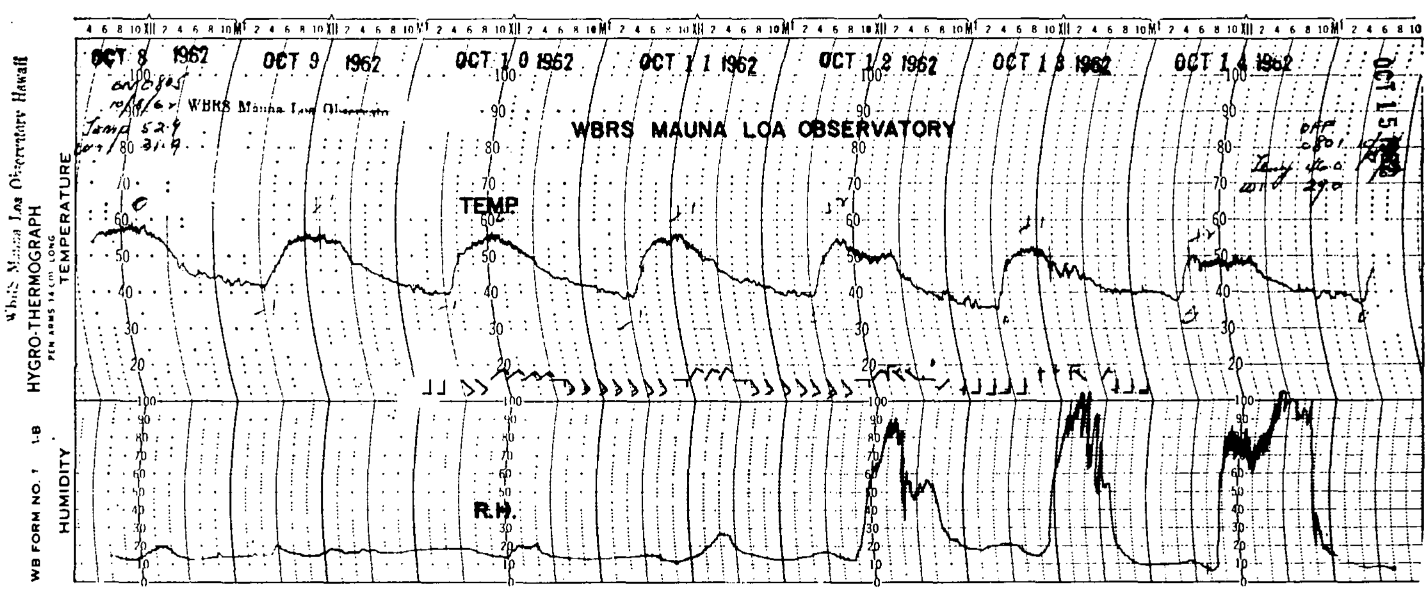


FIGURE 5 —Typical hygrothermograph trace from Mauna Loa Observatory illustrating the afternoon rise in humidity associated with the diurnal upslope flow. The increase, which was scarcely discernible during the daytime northeasterly winds of October 10 and 11, became conspicuous later in the period.

tion winds are characteristically the lightest and most variable of the day.

At Hilo, at the seaward foot of Mauna Loa, the mountain circulation incorporates the coastal land and sea breeze regime, the former appearing as an extension of the downslope currents, and the latter of the east-northeasterly trades. Night and morning winds are usually southwesterly and quite light; and shallow nocturnal ground inversions are common. Onshore breezes spring up by forenoon.

In general, the integrity of the atmospheric envelope about Mauna Loa is least during the day, when convection and turbulent exchange with the surrounding free air are most intense, and greatest at night. The diurnal circulation at MLO was least in evidence on days of strong synoptic winds.

EFFECT OF THE LOCAL AIR MASS

From what has been said it is clear that although taken only a few hours apart (approximately at 08h and 14h), the morning counts at each site were ordinarily made in the descending branch and the afternoon counts in the ascending branch of the mountain circulation, and hence in air of perhaps markedly different sources and recent histories. Against this background, it may be of some interest to re-evaluate the previously noted differences between the MLO and Hilo, morning and afternoon, ice nucleus counts (see particularly table 1 and fig. 3).

The ordinarily low humidity of the downslope nocturnal wind at MLO assigns it to the ambient atmosphere at that station's elevation, so that the relatively low ice nucleus background indicated by the morning counts may also be regarded as that of the free air above the inversion.

It might therefore have been anticipated that afternoon counts at MLO, being made so frequently in air drawn from lower elevations, often demonstrably from below the inversion, would reflect this greater turbidity in correspondingly higher ice nucleus concentrations. That this did not occur during the period under review is clearly indicated by figure 3, and suggests that the source of air in the upslope wind may still have been far above and many miles distant from Hilo and other possible sources of contamination at lower altitudes and near the coast. On the other hand, table 3 does confirm that *in most months*, particularly in the warmer half-year, afternoon counts did exceed those obtained in the morning. In fact, the apparent seasonality in the a.m./p.m. ratio (unfortunately obscured by the lack of afternoon counts in several summer months) may well derive from a corresponding seasonality in the time of onset and the intensity of the upslope flow (and hence, perhaps, of the depth at which the afternoon air originates—a possibility considerably strengthened by Kline's [18] finding, in January 1961, that counts made at MLO *later in the afternoon* than were those in the present study substantially exceeded the morning counts).

TABLE 3.—*Monthly median number of ice crystals in 10 liters at −24° C, Mauna Loa Observatory*

	Number of Days (Paired values, only)*	Median Count	
		a.m.	p.m.
1959			
October	17	2.0	3.5
November	23	3.4	3.6
December	27	3.6	5.0
1960			
January	30	2.9	1.3
February	27	4.6	7.6
March	30	9.4	13.4
April	27	9.2	
May	27	6.2	
June	25	14.6	
July	27	13.2	22.2
August	28	4.4	7.1
September	29	4.2	4.2
October	31	6.2	6.2
November	28	3.4	2.3
December	30	9.3	6.3
1961			
January 1–February 4	35	2.0	**8.0
December 1–February 5, 1962	65	10.6	6.2

*Only days having counts in both a.m. and p.m. were used in computing the medians.
**These observations were obtained (by Kline [18]) somewhat later in the day than were the other afternoon counts.

At Hilo, morning counts were usually obtained beneath a shallow radiation inversion and in a light off-mountain flow—hence within relatively stagnant air which, having spent its recent hours over the island, might be expected to have amassed contaminants of local origin. On the other hand, afternoon counts at Hilo sampled air more freshly arrived from the sea and being mixed by convection through a greater depth of atmosphere.

This contrast between the morning and afternoon air masses at Hilo is reflected in a wide disparity in ice nucleus content. As table 1 shows, the mean of the afternoon counts at Hilo is less than half that of the morning observations, and the median only one-fifth. It would seem reasonable to ascribe these higher morning values (see also fig. 3) directly to the presence of local pollutants, and the lower afternoon counts in part to correspondingly low concentrations of ice nuclei within the onshore flow of relatively uncontaminated marine air. However, some uncertainty still exists concerning the sea as a source of ice nuclei. Kline [18] had found no count increase in onshore winds along the Hilo coast—but see Battan and Riley [1], Bird et al. [7], Brier and Kline [14], and Georgii [17] for typically divergent views. Most investigators presently consider the ice nucleus content of unpolluted marine air to be relatively low.

That daytime turbulence and convection may also have helped lower ice nucleus levels, presumably by dispersing pollutants previously entrapped beneath the nocturnal

TABLE 4.—*Median number of ice crystals in 10 liters at −24° C. in Hilo on weekdays and Sundays, December 1, 1961 to February 5, 1962*

Weekdays		Sundays	
a.m.	p.m.	a.m.	p.m.
150	32	49	24

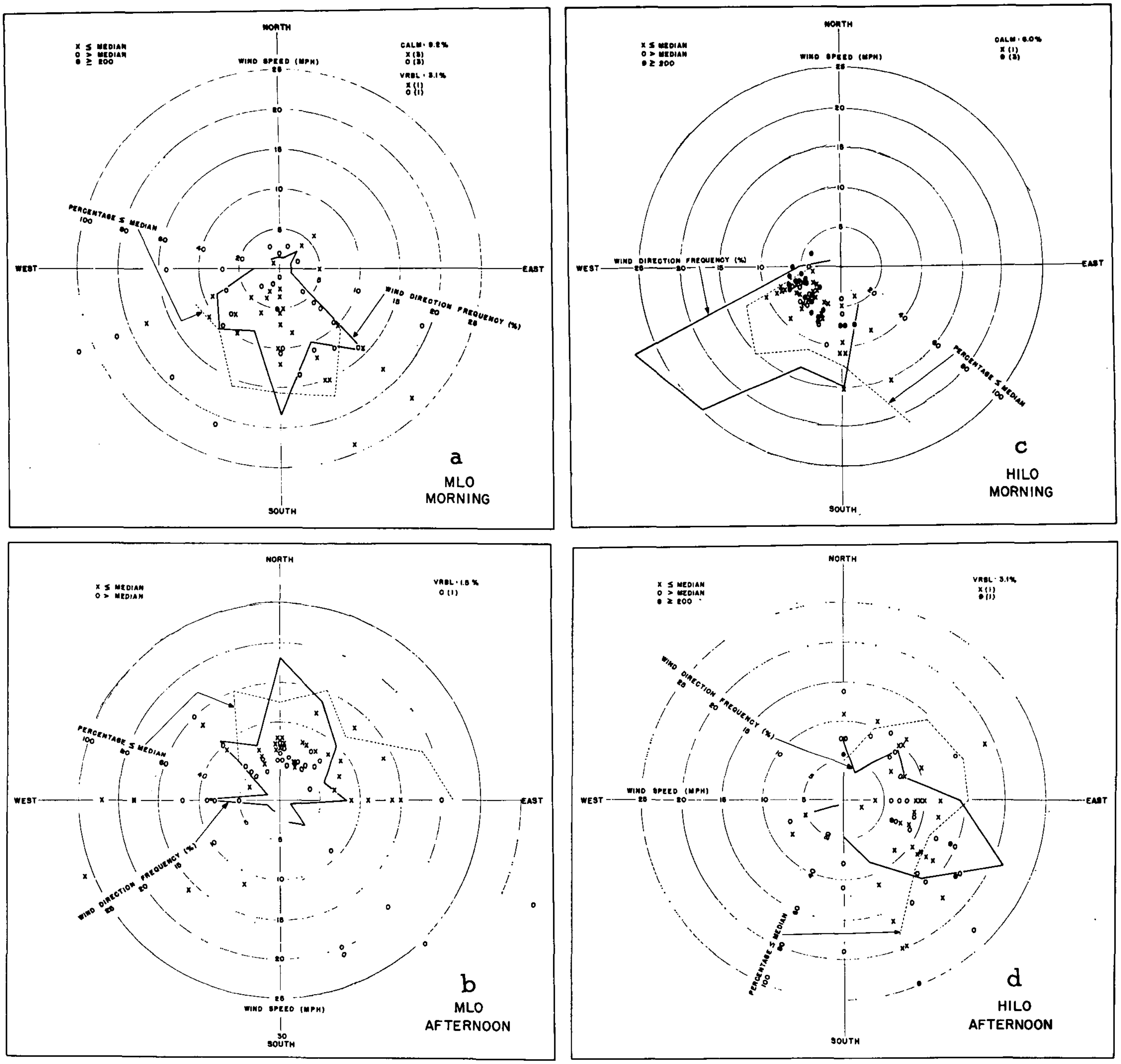


FIGURE 6.—Daily morning and afternoon ice nucleus counts at MLO (a and b) and Hilo (c and d) plotted in terms of their respective medians and as a function of wind speed and direction at the time counts were made. Wind direction frequencies are shown by solid dark lines and the percent of counts equal to or below the median by light dashed lines.

inversion, is supported by the numerous instances when afternoon counts at Hilo, even in air having an overland trajectory, were sharply below morning counts. This would also help account for the previously remarked "alternation" on some successive days between unusually high morning and unusually low afternoon counts.

Conversely, the persistence of high counts throughout other days may have been associated with the persistence of a shallow confining inversion. Thus, December 11—a day with a strong ground inversion on both the morning and afternoon soundings—had a.m./p.m. counts, of respectively, 235 and 200 ice crystals in 10 liters; and January 1, with an unusually low trade inversion (914 mb.) had a.m./p.m. counts of 259 and 422.

Interestingly enough, dust particles counted in Hilo during this period at about the same times of day as the ice nuclei, were also about twice as numerous in the morning as in the afternoon.

That local sources might have been contributing importantly to higher levels in both dust and ice nuclei is suggested by table 4, which shows that both were strikingly lower on Sundays than during the rest of the week. In fact, the lowest morning ice nucleus count at Hilo during the entire period—6 ice crystals/10 liters—was on Monday, December 25, Christmas Day, following a steady decline which began the previous Friday, December 22.

THE EFFECT OF WIND SPEED AND DIRECTION

While it is evident from the foregoing that at both Hilo and Mauna Loa the ice nucleus content of the air seems to be directly responsive to its source and recent history, the specific wind speed and direction at observation time during the period under discussion appeared to exert little further influence on how individual counts compared to others made at the same place and time of day.

This is illustrated by figures 6a to 6d, in which counts have been plotted as a function of wind direction and speed at the time they were made, and in terms of the respective medians in each group of data. Use of the median permitted comparing the actual distribution of counts with expectancy merely by inspection, and this has been done by the dashed lines in each of the figures. Solid lines indicate the directional frequency of the wind, and disclose at once its conspicuous diurnal shift from southerly to northerly at the Observatory (figs. 6a and 6b) and from southwesterly to southeasterly at Hilo (figs. 6c and 6d).

In the morning at Mauna Loa (fig. 6a), relatively low (sub-median) counts appear to have exceeded expectancy most noticeably in southerly (nocturnal downslope) winds of below about 10 m.p.h.

Afternoon counts at Mauna Loa (fig. 6b) were relatively lower in east-northeasterly winds and higher in winds from the southeast, the latter direction—particularly during the day—indicating a synoptic wind and the former the ascent and inflow of air principally from *above* the inversion. In air arriving at MLO from the northeast to northwest—directions commonly associated there with upslope motion—counts were as often above as below the median.

In figure 6c the clustering of points—virtually all of which lie within the southwestern quadrant at 5 to 10 m.p.h.—suggests at once that these morning counts at Hilo could not have been much affected by wind speed or direction.

Afternoon winds at Hilo (fig. 6d), however, are much more diverse, and relatively lower counts are more common in winds from north-northeast to south-southeast, the sector which, as figure 1 shows, opens most directly on the sea, and slightly less frequent with winds from the interior, perhaps implying local pollution sources. But a number of the above-median counts and several of the peaks are also to be found in the onshore winds, so that the possibility of contamination (or of some "natural" source of nuclei) within the freshly arrived marine air cannot be entirely dismissed.

TABLE 5.—*Measures of co-variation between sequences of ice nucleus counts*

Count-sequence Pair	Number of Paired Observations	Number of Matches		(1)	(2)	(3)
		Expected	Observed	χ^2	P_{χ^2}	R_{rank}
Hilo a.m./p.m.	65	32.5	36	0.8	>0.30	0.11
MLO a.m./p.m.	65	32.5	45	9.0	<0.005	0.44
Hilo a.m./MLO a.m.	64	32	32			−0.06
Hilo p.m./MLO p.m.	64	32	28	1.0	>0.30	−0.03
Hilo a.m./MLO p.m.	64	32.5	37	1.3	>0.20	0.20
Hilo p.m./MLO a.m.	63	31.5	32	<0.1	>0.90	−0.29

(1) Computed from discrepancy between observed and expected number of pairs having the same sign of departure from their respective medians. (No correction was made for continuity.)
(2) Probability of obtaining the computed values of chi-square between identical populations. (d.f.=1).
(3) Coefficients of rank correlation between corresponding pairs of ice nucleus counts of ranked count-sequences.

CO-VARIATION AMONG THE ICE NUCLEUS SERIES

It has been shown that the ice nucleus counts described in this report were *in general* obtained under the following rather diverse meteorological circumstances:

1. Morning counts at Hilo within a shallow layer of stagnant marine air containing contamination of local origin;
2. Afternoon counts at Hilo within marine air stirred by turbulence and convection, and replenished at times by fresh onshore winds;
3. Morning counts at MLO in air above the inversion (when present) and representative of the surrounding free atmosphere;
4. Afternoon counts at MLO in air ascending from lower elevations, often from below the trade inversion.

The usual difficulty of ascertaining co-variation among meteorological time series is compounded in these four sets of observations by their wide and rapid fluctuations and by frequent shifts in relative phase among two or more of them. An additional uncertainty arises from the fact that the sampling method employed—a few successive counts made twice daily—is inherently incapable of detecting, except by chance, the shorter-period, often large-amplitude, variations found whenever ice nucleus levels have been continuously monitored [6, 23] (and in short-interval counts at MLO and Hilo, see fig. 7), and could thus easily overlook in some of the count-sequences events fortuitously picked up in others.

In view of the foregoing complications and the contrasting meteorological environments in which the four sets of observations were obtained, an absence of correlation among them might have been less surprising than its presence. Nevertheless, even a casual eye can find a number of concurrences among the plots of figure 2.

To emphasize common trends and variations, the 3-day running means were obtained for each sequence of counts (fig. 8). In general, each of the resulting curves presents a recognizable image of the others, although in places

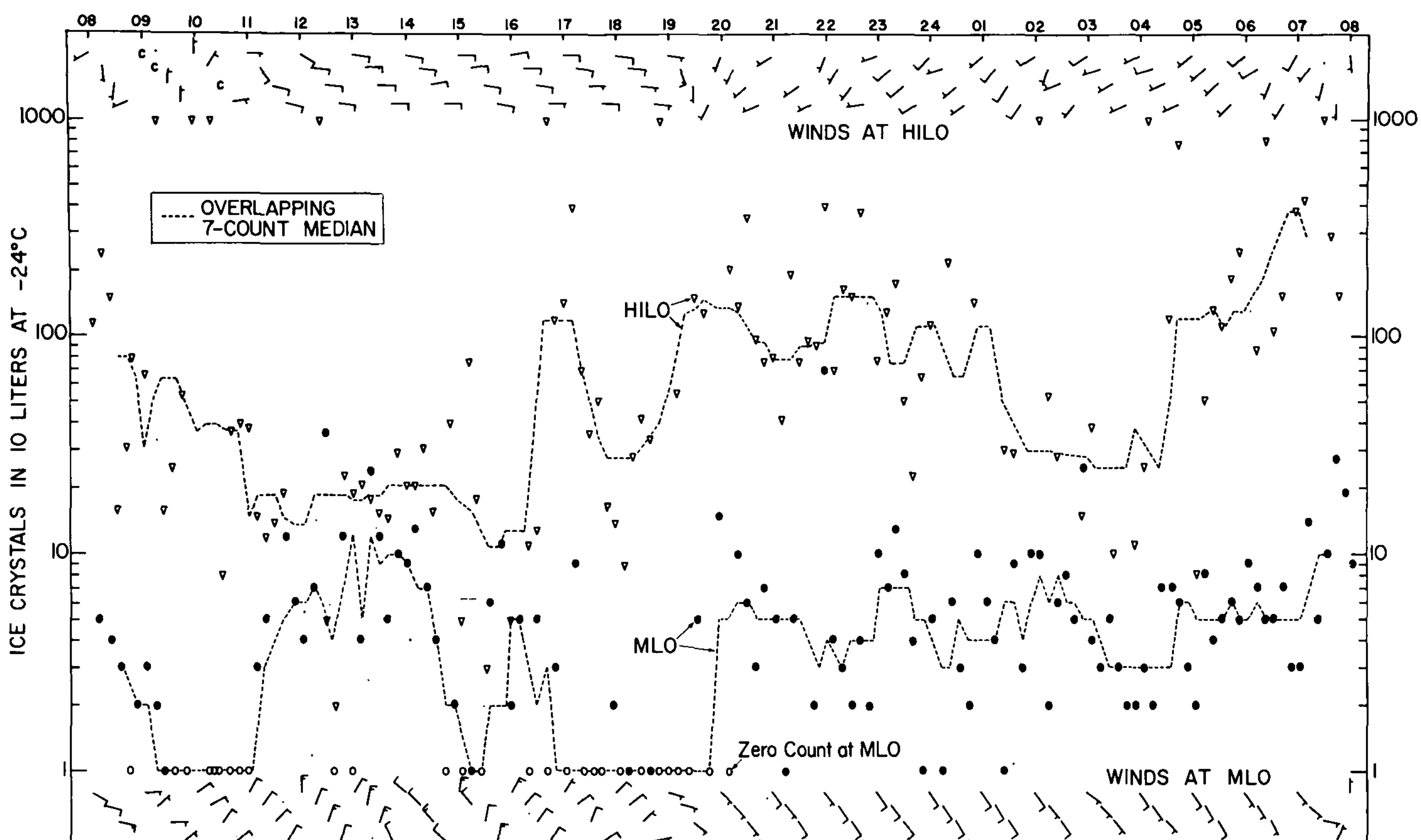


FIGURE 7.—Ice nucleus counts made at frequent intervals at MLO and Hilo throughout the 24 hours beginning about 0800 LST on July 10, 1963. Dashed lines are overlapping 7-count medians.

somewhat distorted in phase and amplitude.[4] It is, however, doubtful that a convincing correspondence could be established for every point and feature; or—as a result of shifting phase relationships among the series—that standard methods would confirm the correlations so evident to the eye.[5]

A number of major as well as lesser correspondences are immediately evident. The most conspicuous include the maxima of December 11–12, 28–30, and January 7–8; the great peaks of January 21–22; the minima of December 24–26 and January 4–5; and the great minimum of January 11–13.

The entire period December 25 to January 25 displays an unmistakable parallelism, and even the very low afternoon counts which persisted at MLO throughout the

[4] Displacements or even reversals in phase may be caused even by use of the running mean: note, for example, that for a.m. counts at MLO between December 28 and January 4, the running mean is directly out of phase with the daily values.

[5] Correlation between the count sequences, taken in pairs, was estimated (by Chi-square) by comparing the actual and expected number of times corresponding counts were both above or both below their respective medians (that is, by matching count-pairs). The expected number of such matches is, of course, $N/2$, where N is the number of pairs.

Coefficients of rank correlation, R_r, were also computed. (Serial correlation within *both time series would tend to increase R_r if the variations of both were in phase.* In the present instance this would be a desirable outcome.)

The summary in table 5 suggests significant correlation only between morning and afternoon counts at MLO. In view of the unmistakable parallelism in the data, this can only confirm the inadequacy of the statistical tests.

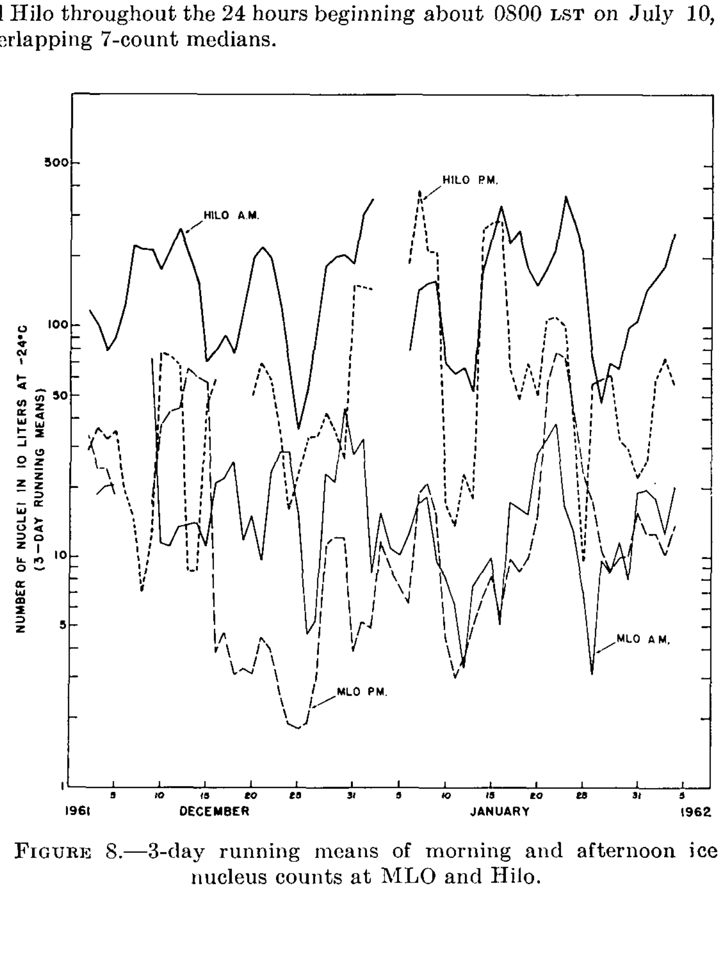


FIGURE 8.—3-day running means of morning and afternoon ice nucleus counts at MLO and Hilo.

latter half of December appear to have varied concurrently with the other counts.

The existence of co-variation among measurements made at two different places and times of day would appear to constitute some validation of them—that is, to suggest that what was being observed was a property of the atmosphere, rather than of the instruments, procedures, or observers. Its existence among measurements made in such diverse meteorological environments would appear further to indicate either a common response to large-scale variations in the atmospheric ice nucleus content, such as might accompany synoptic or extraterrestrial events, or a local exchange of matter between the two sites.

Each of these possibilities will be briefly considered in turn.

Extraterrestrial Events.—The only systematic explanation yet advanced for the occasional major peaks (but not for the background variability) in ice nucleus concentration noted wherever such observations have been made is Bowen's [8] provocative suggestion initially relating rainfall singularities and, later [11], apparently concurrent peaks in cirrus cloudiness and ice nuclei to prior injections of meteoritic material into the high atmosphere.

It has been pointed out, for example by Fletcher [16], that the abruptness of these geophysical singularities and the fact that they center near the same calendar dates for events ranging over great distances and in altitude from 10 km. (cirrus) to sea level (ice nuclei) imply that the allegedly responsible material undergoes minimal dispersion during its descent from the upper atmosphere, reaches the tropopause in sharp pulses, and then very rapidly diffuses throughout the troposphere.[6]

Hence, although gravitational settling in still air would produce noticeable lags between arrival times at MLO (3.4 km.) and Hilo (sea level) (about 2.4 days for spherical iron particles of radius 8μ and density 8 gm. cm.$^{-3}$, and double that for stony particles of the same size), the foregoing hypothesis implies that nuclei descending from aloft could be expected to reach both places at about the same time.

In actuality, up- and downslope motions along Mauna Loa and the intervening trade inversion could make the MLO-Hilo lag highly variable. In any event, no consistent evidence of a lag can be found in the data.

The isolation of Mauna Loa Observatory by elevation, insularity, and—particularly during the night and morning—by the inversion and downslope winds from much of the turbidity of the lower atmosphere suggests it as especially suited to the detection of influxes of nuclei from higher in the atmosphere. Further, although such nuclei might, because of dispersion, be no more numerous at MLO than at lower elevations, they would ordinarily be *proportionately* more abundant at the Observatory, and hence more conspicuous against its much lower background count than when superimposed upon the already higher and more variable counts at Hilo.

But even at MLO the detection of an extraterrestrial component is complicated not only by the unsteadiness of the background count and the inadequate time-resolving power of the counter, but also by uncertainties stemming from differences among authoritative lists of meteor shower dates, the discovery of new showers [15], the persistence of most showers through several or more days, variations of shower-singularity lags (e.g., particle descent-times from the upper atmosphere) with area and period of record (Bowen [11], for example, uses 28 to 31 days, Maruyama [23] finds the 28-day lag most significant, Dimitriev and Chile [15] indicate lags of 28 to 35 days). Since *minor* peaks or dips in the counts are themselves often only a day or two apart,[7] and major anomalies of each sign occur at least several times a month, the permissible latitudes in dating could easily produce or avoid coincidences, and thus make questionable any but the most striking and consistent effects of extraterrestrial nuclei on the counts.

The most conspicuous correspondence appears to be that between the Ursids [8] and the high counts of January 22–23, when all four of the Hawaiian ice nucleus series peaked sharply. These dates mark also prominent singularities in rainfall over the world [8] and in the U.S.S.R. [15], in cirrus frequency [3], and in ice nucleus measurements made in various localities in 1954 to 1958 [20].

However, during the winter of 1959–60 rainfall in the United States and ice nucleus levels had deep *minima* at those dates (from fig. 11 in [19]), which suggests either that the earlier association is fortuitous or that over limited areas and periods it may be overridden by other factors. (Brier [12], for example, finds a long-term correlation between world and United States rainfall.)

Other less conspicuous concurrences may also be intimated by the data. It would, for example, be easy to see in the generally elevated counts of about December 28 to January 8 the influence of the similarly prolonged Bielids I, and in the included sharp peak of January 8 (evident in all four series) that of the Bielids II. The Quadrantids (February 1) also occur during relatively high counts, but the rise had begun several days earlier; while the Geminids, which—like the Ursids—are strongly associated with other geophysical singularities, can be regarded as related either to the deep minimum of January 11–13 in the Hawaiian counts, or to the beginning of a steep rise which culminated in the high levels of January 15.

[6] This apparent resistance to dispersion above the tropopause by gravitational settling, turbulence, and large-scale vertical exchange remains to many one of the most troublesome aspects of the theory; nor has the equally abrupt termination of the effect been clearly accounted for.

[7] The minimum period between successive counts higher than either adjacent count is 2 days, and the maximum number of such "peaks" in these ice nucleus count-sequences, 32. Morning counts at MLO had 24 such peaks, of which 12 occurred at the minimum interval (2 days) and 7 at 3-day intervals. Of the 20 afternoon peaks, 6 occurred at 2-day intervals, and 6 others at 3-day intervals. Major peaks were, of course, less frequent.

[8] Dates given for meteor showers are those of the occurrence plus 30 days, principally as listed by Bowen [9, 10].

Exchange of Terrestrial Materials.—During recent volcanic eruptions on the Island of Hawaii, effluents sporadically emitted well below the inversion (e.g., near sea level and at 3500 ft.) and at a distance of 25 to 50 mi. reached MLO within one to several days, the lag and concentration appearing to depend on the strength and location of the source, ambient wind, inversion strength and height, and the intensity of the daytime upslope flow. Residual volcanic smoke and haze visible some distance below the Observatory in early mornings frequently moved in by afternoon or before [29].

Similarly, the possibility can not be lightly dismissed that co-variation between the Hilo and MLO ice nucleus counts could arise simply from an exchange of terrestrial material: that is, from a transport to one site of aerosols having their source nearer the other. The generally higher ice nucleus levels found at Hilo, and the prevailingly upward movement of air along Mauna Loa during the day, suggest that, particularly then, ice nuclei of local origin are more likely to move from Hilo to MLO than the reverse; at night, the absence of convection and the resistance offered by compressional heating and the inversion (when present) to the descent of the radiatively-cooled slope currents, would appear to minimize nocturnal exchange between the two sites.

On the exchange hypothesis, one would expect the Hilo and MLO counts to be more closely related when the mountain circulation is well developed than when it is not and during deep vertical mixing, as indicated by the absence of the trade inversion and the presence of a deep moist layer in the free air. That these expectations are not sustained by a close examination of the data may be attributed to the complexities (of sampling, count fluctuations, etc.) discussed earlier, and to such others as the following.

It was pointed out earlier that the contaminants which may be responsible for Hilo's high *morning* counts are ordinarily trapped beneath a shallow radiation inversion over the lower slopes of Mauna Loa until after sunrise when they are dispersed by convection and swept inland and upslope by the onshore and mountain winds. If these pollutants reach MLO at all, they presumably do so—as the volcanic observations suggest—after intervals of variable, but unknown, length. Variations in this interval and in the degree of dilution experienced en route could easily have a greater effect on ice nucleus concentrations subsequently measured at MLO than might the initial abundance as reflected in the morning ice nucleus levels at Hilo; so that the latter can be regarded as little more than a crude index of potential availability. It would, therefore, seem likely that the increased ice nucleus levels sometimes observed at MLO in later afternoon represent not specifically the concentration in the Hilo area earlier that day, but rather the generally higher turbidity of the air from lower elevations, particularly from below the inversion.

Effects of Synoptic Change.—The most consistent response of ice nucleus levels to specific meteorological

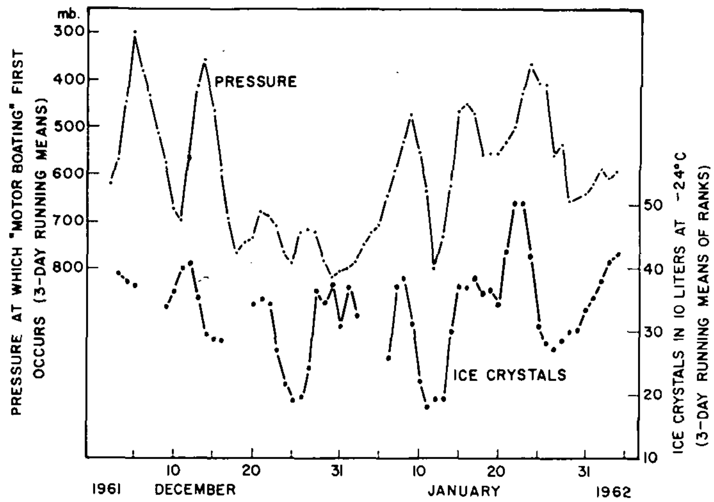


FIGURE 9.—Major oscillations in the number of ice crystals in 10 liters at −24°C. at MLO and Hilo, as shown by 3-day running means of combined ranks, appear to vary with depth of moist layer (pressure at which "motorboating" first occurs in the Hilo radiosonde observations) as an index of synoptic change.

events was reported by Kline [19], who noted pronounced changes in the spectrum of nuclei number vs. temperature with frontal passages in the Washington, D.C., area. Following each of nine such "major cold front passages," ice nucleus concentrations throughout the range −10° to −25°C. (and in seven instances to −30°C.) fell up to two orders of magnitude below their former values. Whether these differences are intrinsic to the pre- and post-frontal air masses, or represented a reduction in contamination at the observing site by other factors associated with the front—e.g., the wind shift or vertical exchange—is not, however, readily determinable. Rau [32] described counts as being *higher* in the polar air, while others (e.g., Mossop et al. [27], Bigg [2]) found no synoptic relationships worthy of note.

The variations which in the Hawaii data would a priori seem most likely to have been associated with synoptic changes are those which appear strongly in all four series: for example, the peaks of January 7–8 and 21–23, and the minimum of January 11–13. The last of these occurred during light trades, while the peaks of January 8 and 22 may have been associated with surface fronts or shear lines which appear to have passed Hawaii.[9] On January 8 the increase lasted only a day, counts being much higher with or immediately preceding the front than either before or after; the high counts which began on January 22 lasted for several days. On both these occasions the increase at MLO, preceded by a day at Hilo, is somewhat discounted by the fact that although the possibility that nuclei from higher aloft may have been involved, counts at the Observatory were less than one-tenth those at Hilo.

The interesting period of December 15–28, when the

[9] The indefiniteness of this statement is deliberate, and is meant to reflect the uncertainties in synoptic analysis stemming from data sparsity in the central Pacific near Hawaii and the dimished frontal contrasts in mid-ocean.

afternoon ice nucleus level at MLO remained remarkably low for 14 successive days, was dominated throughout by a large Pacific High. Although on a number of these days the diurnal cycle in humidity and wind at the Observatory suggested a well-developed mountain circulation, air reaching MLO even during the afternoon appears to have been drawn from above the inversion, although from elevations well below the station. Morning counts at MLO included no peaks, although several were above the median; at Hilo a number of major peaks occurred. Neither at the surface nor aloft, however, was there any evidence of synoptic perturbations with which these fluctuations in count might have been associated.

The broad plateau of relatively high counts from January 15 to 20 and the minimum of January 3 to 5 both occurred with southerly winds which, during each interval, became at times strong enough to require small craft warnings. Similarly, trades ranging from gentle to fresh prevailed from about December 9 to 29, a period marked by wide oscillations in the counts.

Despite the relative infrequency and diffuseness of fronts in the central Pacific and of continental-maritime contrasts (air from whatever source will have traversed thousands of miles of open ocean), air mass changes in Hawaii do occur. These, however, usually involve not the replacement of one air mass by another of different properties, but rather a vertical redistribution of heat and water vapor—the lapse rates of temperature and humidity often being profoundly altered by the convergence and divergence which accompany easterly waves, shear lines, and other large-scale synoptic entities, as well as by other events too small or subtle to register clearly on the sparse surface or upper-level observational networks. These air mass changes are reflected most clearly in the height and strength of the trade inversion (including its extension and reappearance) and in the depth of the moist layer.

As an objective index of these changes, the depth of the moist layer on the daily 0000 GMT (1400 LST) Hilo soundings has been plotted in figure 9 in terms of the pressure at which "motor-boating" was first reported, together with the ice nucleus counts expressed as ranks (obtained by arraying each set of observations) and summed over the four series. The correspondence between the two curves is at once evident, the count rising and falling more or less in unison with the deepening and thinning of the moist layer. Hence, although a close study of the surface and upper-level synoptic charts discloses no unmistakable and consistent response to large-scale stimuli by either the individual or averaged daily counts, it would nevertheless appear from the foregoing that the major oscillations in ice nucleus concentration at MLO and Hilo may be synoptically induced.

DIURNAL VARIATION IN ICE NUCLEI AT MLO AND HILO

Throughout this report various of the differences between, and diurnal variations in, ice nucleus levels at MLO and Hilo, found in the twice-daily counts of December 1961 to February 1962 and previously, have been interpreted in terms of the changing atmospheric environments induced by the diurnal reversals of wind and other aspects of the mountain circulation about Mauna Loa.

To validate these inferences it was thought desirable, despite the difficulties involved,[10] to investigate the diurnal variation of ice nucleus concentrations at MLO and Hilo more directly by measurements made concurrently and at frequent intervals throughout at least a single day. This was done for the 24 hr. beginning at 0800 LST on July 10, 1963, with the expansion chambers, procedures, and observers employed in securing the earlier data. Counts were made at approximately 10-min. intervals, a total of 135 being obtained at MLO and 113 at Hilo.[11]

The resulting observations are shown in figure 7, together with the wind speed and direction at the time of each count. Perhaps most immediately striking is a variability as great as that previously noted between successive days, with counts made only minutes apart differing at times by an order of magnitude or more (compare with fig. 2). (Comparable variabilities have also been found elsewhere by continuously recording ice nucleus counters [6, 23].)

For the most part, ice nucleus levels at MLO ran well below those at Hilo. For example, as table 6 shows, 21 percent of the counts at MLO yielded no ice crystals at all, and 85 percent yielded fewer than 10 (in 10 liters at $-24°$C.); at Hilo only 7 percent of the observations were under 10 ice crystals and 38 percent exceeded 100.

On the whole—again as had been anticipated—counts at Hilo were higher during the night and early morning (presumably because of overland air trajectories, light winds, and the retention of contaminants below shallow ground inversions) and lowest in the hours when daytime convective mixing and the onshore (easterly) winds would be jointly most effective in reducing pollutant levels. At MLO the expected afternoon increase in ice nuclei was clearly evident, delineating the influx of more turbid air from beneath the inversion. Also, counts at the Observatory tended to be higher in the more northerly than in the more easterly winds—sustaining an earlier surmise, derived from their lower dew points, that the latter represent air principally from above, rather than from below, the trade inversion.

Despite their usual wide disparity, the MLO and Hilo counts did overlap at times, most noticeably from about noon to 1600 LST of July 10, although even then the MLO median lay well below Hilo's. But, as was stated earlier, this probably implies not an actual exchange of material between the two sites, but only that the dispersion of contaminants at Hilo and the arrival of sub-inversion air at MLO occur during the same portion of the day.

[10] Since a count requires about 10 min., both instruments had to be operated continuously by relays of observers.

[11] "Count" in this 24-hr. run means a single determination (the number of ice crystals formed in a purging-expansion cycle), not the average of five successive such determinations, as it does elsewhere in this report.

TABLE 6.—*Frequency distribution of ice crystal counts at MLO and Hilo during short-interval observations, July 10–11, 1963*

Number of Ice Crystals*	Percent of Observations in Given Range	
	Mauna Loa Observatory	Hilo
0	20.7	
1–9	63.6	7.1
10–19	11.9	16.8
20–49	3.0	20.4
50–99	0.8	17.7
100–199		18.6
200–999		11.5
999		8.0

*In 10 liters at −24°C.

5. CONCLUSIONS AND CONJECTURES

In all likelihood air entering the expansion chambers at Mauna Loa and Hilo contained a diversity of particulate and nonparticulate matter derived initially from sources near and remote and from a wide range of natural and human activities. Some may have been of extraterrestrial origin.

Nothing is known concerning the identity or size distribution of the components, their physical and chemical interactions in the atmosphere or within the expansion chamber itself, or other factors which could have influenced their nucleating efficiencies. There would, however, seem no more reason for assuming constancy in the composition and state of this material than in its abundance, so that the variations in ice nucleus count at MLO and Hilo may have arisen from changes not only in the size of the population but also in its nature, origin, and history.

In short, the only property investigated in the present study was the tendency for countable ice crystals to form under the highly specific conditions described, and all conclusions concerning their number and variations at MLO and Hilo must be viewed against these limitations.

The data quite explicitly show that during the period under review:

1. Ice nucleus counts at both Mauna Loa Observatory and Hilo fluctuated widely, not only from day to day (fig. 2), but also by as much as an order of magnitude within minutes (fig. 7).

2. Background ice nucleus levels at Hilo ran well above those at MLO, and peaks tended to be higher and more numerous (figs. 2 and 3 and table 1).

3. At Hilo, morning counts averaged higher than afternoon counts and peaks were more common. At MLO, counts made in later afternoon tended to exceed those made in the morning.

The data also contain the following *implications:*

1. The Hilo ice nuclei appear ordinarily to have originated in the lower atmosphere—specifically in the air below the trade inversion. Morning counts were higher and more frequently peaked presumably because they were taken within relatively stagnant air containing pollutants of local origin. Afternoon counts appear, in general, to represent the ice nucleus content of "clean" sub-inversion marine air in the Hawaiian area, but may be affected by the presence of contaminants in greater dilution.

2. At Mauna Loa Observatory, morning counts often reflect the ice nucleus content of the ambient free air, while afternoon counts are typically made within air from lower elevations, frequently from below the trade inversion.

3. MLO's low ice nucleus levels (low relative not only to Hilo but also to other isolated areas) are attributable to its insulation by distance, elevation, and the trade inversion from the underlying and more turbid atmosphere.

4. The four ice nucleus sequences, composed respectively of counts made each morning and afternoon at MLO and Hilo, appear to be clearly correlated (fig. 8). So marked a parallelism in observations obtained within such diverse meteorological environments suggests a common response to large-scale or local influences, but also supports the validity of the measurements themselves.

5. The major oscillations in ice nucleus levels at MLO and Hilo appear to accompany air mass changes or other synoptic events as manifested not in the grosser features of the synoptic chart, but in the deepening and thinning of the moist layer (fig. 9).

6. There is no conclusive evidence for (or against) the Bowen hypothesis.

Although the observations described in this report were not intended to elucidate any of the more fundamental questions concerning the atmospheric ice nuclei, implications do arise for at least one of them, for example, the source and vertical distribution of the ice nuclei counted at MLO and Hilo during the period under review.

It has already been pointed out that the low background count at MLO relative to that at Hilo (see again fig. 3) clearly implies, first, that the ice nuclei observed at Hilo did not *ordinarily and in major part* come from aloft, but rather from the lower atmosphere and, further, that the number of ice nuclei decreases with height in the Hawaiian area.

The latter inference would on its face clearly disagree with a number of observations made by aircraft and balloons which suggest ice nucleus concentrations some distance above the earth are greater than near its surface. Telford [34], for example, reported that counts at 25,000 ft. (in Australia) ran well above those at sea level (the latter values were, however, only assumed; no actual concurrent counts are cited). Even more striking, Murgatroyd and Garrod [28] found much higher counts above than below "a well-marked temperature inversion" in southern England, and from this and from condensation nucleus counts made during the ascents to flight altitude concluded that the earth's surface was not the principal source of the ice nuclei encountered aloft. Although one may assume that their measurements were made in uncontaminated air (the authors give no details on the track of the sampling aircraft or the altitudes of its flights or of the inversion), the number of samples was so small (five), the disparity between levels so great (a factor of

10^4) and the ice nucleus concentration above the inversion so high (10,000 per liter at −24°C.) as to invite question. More recently, large numbers of ice nuclei, some of them active at temperatures as high as −10°C., have been found at altitudes of 20 km. and above [4]; so that there appears to be appreciable evidence (not all of it, however, unequivocal) for an abundance of ice nuclei at high altitudes and for their increase in number with height above sea level.

The generality of the Hawaiian data and their relevance to vertical ice nucleus gradients elsewhere in the free air would seem to hinge on whether the observed differences between MLO and Hilo represent anything more than a local contrast between turbid air below the trade inversion and cleaner air above. Did the Hilo counts reflect concentrations over the nearby open sea (were they "naturally occurring" nuclei?) or did they involve, even during the afternoon, contamination from sources (cane fires, sugar mills, vehicular traffic) near Hilo and elsewhere on the island of Hawaii? What, in fact, should be taken as the ice nucleus content of the uncontaminated marine air below the trade inversion?—the lowest counts in the onshore winds (fewer than 5 ice crystals in 10 liters at −24°C.)? or, perhaps more realistically, the median of approximately 40 noted by Kline [19] on the coast near Hilo? or of about 28 obtained during the afternoon in the present study a few hundred yards inshore in air deeply stirred by turbulence and convection, and hence presumably containing local contaminants only in high dilution?

The significance of the choice for the vertical gradient of ice nuclei in the Hawaiian area is at once evident. Thus, the lowest of these values (5 nuclei in 10 liters) for Hilo would imply no important difference in ice nucleus content between the air near the sea and that at the elevation at MLO, while the higher counts, which clearly exceed those at the Observatory, would indicate a marked decrease with altitude from the surface to approximately 3.5 km.

That all these values may simply express conditions through the trade inversion is suggested by six days on which that inversion was absent and moisture to great heights gave evidence of deep mixing. Counts at MLO ran well above the median and on four of the afternoons exceeded counts at Hilo. Since dust levels at MLO and afternoon ice nuclei counts at Hilo were both low on those days, it seems less likely that the higher counts at MLO stemmed from the upward transport of aerosols otherwise confined beneath the trade inversion than that they reflected the prevailing synoptic conditions. Hence, the atmospheric ice nuclei in the Hawaiian area may ordinarily decrease in number upward through the trade inversion and remain more constant or increase slightly with height when it is absent and deep mixing occurs.

In view of the foregoing, the case for a source near sea level as against higher in the atmosphere for a major portion of the Hilo ice nuclei, and for at least some of those counted at MLO, would appear to rest chiefly on the following:

(1) Ice nucleus counts at 3.5 km. (MLO) were, for an extended period, extraordinarily low relative to those at sea level in the Hawaiian area as well as in other isolated localities.

(2) Ice nuclei in appreciable numbers could not have consistently reached Hilo from aloft without being detected at MLO during their descent.

(3) Afternoon ice nucleus levels at MLO frequently rose during the influx of air from lower elevations.

(4) Variations in ice nuclei at MLO, while clearly correlated with those at Hilo, were in the main too small by an order of magnitude to have produced them; in fact, the lesser frequency and usually smaller amplitude of the MLO peaks suggest that variations in the extraterrestrial (or other high-atmosphere) component might at best make only a minor contribution to the Hilo oscillations.

This does not preclude the possibility that influxes of nuclei from the upper air could on occasion have been responsible for specific fluctuations on both sites, but it would be reasonable then to expect the excess of counts above the background level to be at least as great at MLO as at Hilo.

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